

Amendments to the Specification:

Page 1, lines 2 -7, please replace the paragraph with the following amended paragraph:

This is a Divisional of Application Serial No. 09/506,160 filed February 17, 2000, now Patent No. 6,624,420. This invention relates to a single crystal as scintillating detector for gamma ray or similar high energy radiation which single crystal is composed of Cerium doped Lutetium Yttrium orthosilicate (LYSO) with the general composition of $\text{Ce}_{2x}(\text{Lu}_{1-y}\text{Y}_y)_{2(1-x)}\text{SiO}_5$ where $x = 0.0001$ to 0.02 and $y = 0.0001$ to 1.9999 and claims priority based on U.S. Provisional Application Serial No. 60/120,500 filed Feb. 18, 1999.

Page 2, lines 7 – 17, please replace the paragraph with the following amended paragraph:

Since the turn of this century; there are a large number of crystals proposed for potential scintillating applications. Even though they do show scintillating properties, none of them has all the right properties. The common problems are low light yield, physical weakness and ~~difficult to produce~~ difficulty in producing large size high quality single crystals. Despite the problems, a number of them have found applications in physics, chemistry, geology and medicine. One common feature of all these usable crystals is that they are the only crystals which can be produced in large size and high quality by an industrial manufacturing process with reasonable cost. This has proven to be the most important factor than the details of scintillating properties to be considered as a viable scintillator material. The specific examples include bismuth germanate ((BGO) which is $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ $\text{Bi}_4\text{Ge}_3\text{O}_{12}$), cerium doped gadolinium orthosilicate ((GSO) which is Gd_2SiO_5) and cerium doped lutetium orthosilicate ((LSO) which is Lu_2SiO_5).

Page 3, lines 2 – 10, please replace the paragraph with the following amended paragraph:

In early 80's, the Ce doped GSO crystal was disclosed as a scintillator material. It has adequate density (6.71 g/cm^3) and is also non-hydroscopic. The light yield is 20% of that obtained with NaI(Tl) with a much faster decay time (60 ns). Even though GSO crystals over 80 mm diameter have been produced, the crystal has not yet made it in the PET ~~marker~~ market

because of a strong cleavage plane. It is very difficult to cut and polishing polish the crystal into any specific shape without the risk of fracturing of the entire crystal. Another unexpected problem is the high thermal neutron capture cross-section (49,000 barns) of the gadolinium. It will interfere with the gamma rays generated by neutron irradiation source. However, since there is no neutron source involved in the PET process, gadolinium containing GSO is not a problem.

Page 4, lines 9 to page 5, line 1, please replace the paragraph with the following amended paragraph:

At present, the scintillation process has been well accepted and used in many applications. The basic mechanism is also reasonably well understood. It is generally accepted that the basic scintillation process involves three steps: (1) the absorption of the incident high energy radiation and the conversion into a large number of low energy (a few multiples of the band gap energy) electrons and hole pairs; (2) transfer the electron-hole recombination energy to the luminescence centers before its loss to ~~multi phonon~~ multi-photon relaxation processes; and, (3) the radiative emission of the transferred energy. In other word, the scintillation efficiency (E) can be expressed as:

$$E = \beta \times S \times Q$$

where β is the conversion efficiency, S is the transfer efficiency and Q is quantum efficiency of the radiation centers. Despite the understanding of scintillating mechanism based on the known materials, there is still lack of any good model which has the capability to predict the scintillating behavior of a specific compound. The quantum efficiency of an emission center can be predicted and tested optically; however, neither the total number of electron-hole pairs generated by an incident gamma ray radiation nor the transfer efficiency can be predicted or independently tested. In the end, the only way to confirm the scintillating behavior of a compound is to make and then test it.

Page 6, line 21 to page 7, line 4, replace the paragraph with the following amended paragraph:

The embodiment of this invention is to design a new crystal which can eliminate most of the problems of LSO crystal without sacrifice sacrificing the scintillating properties. Our initial

motivation is to reduce the growth temperature of LSO single crystals. It is a very difficult task to maintain the operation at such high temperature for long period of time (~~<1 week~~) (greater than 1 week). Since YSO has lower melting temperature near 2070 °C, we are seeking the possibility to find an intermediate composition (or LYSO composition) which may melt at lower temperature to ease the growth process. We also want to minimize the yttrium content to retain the LSO scintillating properties.

Page 7, lines 5 – 16, please replace the paragraph with the following amended paragraph:

Since there is no known published phase diagram between YSO and LSO, the phase relationship of the intermediate composition is not available. We speculate the melting and crystallization behavior of the intermediate LYSO crystal composition based on two assumptions. First, since both YSO and LSO have the same crystallographic structure and the ionic size of yttrium 3+ ion (0.090 nm) and lutetium 3+ ion (0.088 nm) are very similar, we assume that there is a 100% miscibility between the ~~m'o~~ two compositions. In other ~~word~~ words, it is possible to make any intermediate composition LYSO crystals without worry about phase separation or formation of new compounds. Second, since YSO has lower melting temperature, based on the model of ideal solid solution, it is expected that all the intermediate compositions will have lower melting temperature similar to the classic example of fosterite (Mg_2SiO_4) - fayalite (Fe_2SiO_4) pseudo-binary phase relations even though the exact position of the solidus and liquidus lines are not known.

Page 7, line 17 to page 8, line 3, please replace the paragraph with the following amended paragraph:

In order to understand the melting and crystallization behavior, four intermediate LYSO charge compositions were prepared. The compositions were: $Ce_{0.002}(Lu_{0.7}Y_{0.3})_{1.998}Si_2O_5$ designated (70% LYSO); $Ce_{0.002}(Lu_{0.5}Y_{0.5})_{1.998}Si_2O_5$ designated (50% LYSO); $Ce_{0.002}(Lu_{0.3}Y_{0.7})_{1.998}Si_2O_5$ $Ce_{0.002}(Lu_{0.3}Y_{0.7})_{1.998}Si_2O_5$ designated (30% LYSO); and, $Ce_{0.002}(Lu_{0.15}Y_{0.85})_{1.998}Si_2O_5$ $Ce_{0.002}(Lu_{0.15}Y_{0.85})_{1.998}Si_2O_5$ designated as (15% LYSO). The percentage refers to the fraction of the lutetium in the crystal. A pure LSO charge was also prepared to be processes processed in a

similar way as a reference for direct comparison. To make sure that the property comparison is meaningful, all the LYSO crystal preparation procedures are identical. The same total number of moles of chemicals in each case were used so that the finished crystals are near identical in size. To minimize the repetition, the 70% LYSO composition is hereafter set forth as the example to illustrate the preparation for all examples:

Page 9, lines 8 – 14, please replace the paragraph with the following amended paragraph:

To evaluate the scintillating properties, two 10 mm slabs were cut from each ~~erystals~~ crystal, one ~~form~~ from the top of the crystal and one from the bottom right above the defect region. It has been found that the scintillating light yield of any LSO crystal decreases systematically from the top to the bottom of the boule. The relative light yield can vary by as much as a factor of 2. It appears that this is due to the impurities in the lutetium oxide (Lu_2O_3) source material. Since the LYSO also uses the same ~~$\text{Lu}_2\theta_3$~~ Lu_2O_3 starting material, the LYSO will also show the same behavior.

Page 10, lines 15 – 20, please replace the paragraph with the following amended paragraph:

~~Fist First~~, the top portion of each crystal has the best scintillating value n since the crystallization process has been found to be a purification process. ~~The first (top) portion~~ The result has many important implications. ~~of a crystal will have the least impurity content and thus the best performance.~~ This result has many important implications. ~~The top portion of the crystal will have the least impurity content and thus the best performance.~~ It is interesting to ~~notee~~ note that the pure LSO crystal has produced light yield of 93% of that of NaI(Tl). This is ~~significant significantly~~ significantly higher than the published result of 75%. This value may approach to the ultimate scintillating power for LSO.

Page 11, line 14 to page 12, line 2, please replace the paragraph with the following amended paragraph:

In addition to the advantage directly observed from the light yield measurements, LYSO also resolves other problems associated with pure LSO. First, the growth temperature of LYSO is lower than that of pure LSO by approximately ~~100~~ 100 °C which is very significant in high temperature processes. Since the radiation heat loss is proportional to the 4th power of temperature (or T_n) the high temperature insulation and iridium crucible will last longer. Second, substituting yttrium will reduce proportionally the trace concentration of the naturally radioactive Lu¹⁷⁶ isotope without sacrificing the net light yield. This will, in effect, reduce the background noise of the detector. Third, both the cost and purity of the ~~Lu₂O₃~~ Lu₂O₃ starting material is a serious issue. Thus, Yttrium substitution will reduce the cost and improve the uniformity of scintillating efficiency for large single crystal plates. Further, since a low index of refraction is preferred for scintillating crystals to reduce the effect of total internal reflection, the substitution of yttrium reduces the already low value of the index of refraction of LSO.